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RECENT FLAME AND COMBUSTION STUDIES BY
Ya. B. ZEL'DOVICH AND OTHER
SOVIET SCIENTISTS

[Note: Scientists of the Institute of Chemical Physics (of the Department of Chemical Sciences, Academy of Sciences of the USSR) in Moscow have been engaged for some time in theoretical and experimental flame and combustion research. In addition to exhibiting a marked degree of general concentration in this field, researchers from the Institute of Chemical Physics and from collaborating institutes (Institute of Physical Chemistry imeni Pisarzhevskiy of the Ukrainian Academy of Sciences, the Institute of Physical Chemistry imeni L. Ya. Karpov of the Academy of Sciences of the USSR, and others) have specialized to a great extent also in investigating the various aspects of these phenomena.

Exemplary as both a prolific and at the same time a more or less specialized contributor on the subject is Doctor of Physical and Mathematical Sciences Ya. B. Zel'dovich, Corresponding Member of the Academy of Sciences of the USSR who was awarded the Stalin Prize, Second Class, for his work on the theory of combustion and detonation of gases which appeared in 1941 and 1942.

Since abstracts and in some cases translations of his previous publications have been made, attention is now directed to his most recent available paper (1), written this time with N. N. Simonov.]

This latter investigation (1) gives the theoretical relationship of the minimum energy required for the spark ignition of an explosive mixture to the physical properties of the mixture, the products of the combustion, and to the velocity of the flame. The velocity of the

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flame denotes the nature of the chemical reaction in the flame.

The ignition of a CO / O_2 mixture was investigated experimentally with respect to the percentage of hydrogen contained in the mixture (affecting the rate of the flame), and with respect to the pressure of the mixture. The data obtained experimentally conform satisfactorily to the theoretical relationships originally proposed, and make it possible to state the equation $R = 3\delta$, where R is the radius of a sphere which can be heated with a spark to the combustion temperature, and where δ is the width of the heated zone in the flame. This equation makes it possible to compute the spark energy necessary for the inflammation of a mixture whose rate of combustion is known.

A detailed review of this article will be given farther on. Here, two additional articles (2,3) by members of the Institute of Chemical Physics will be considered. (Four articles in all comprise the total of materials published on all subjects in available periodical literature by the Institute of Chemical Physics in the period September-December, 1949.)

The first (2) of the two additional studies was written by Z. K. Mayzus, V. G. Markovich, and M. B. Neyman, and deals with the decomposition of butane with the addition of diethyl peroxide. This mixture was observed by the static method at 100 and 200 mm mercury pressure and by the dynamic method, at atmospheric pressure and at temperatures of from 350 to 530°C, and they concluded that the addition of diethyl

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peroxide (3%) does not increase the rate of decomposition of butane.

The co-authors of the second work, L. I. Avramenko and R. V. Lorentso [3], have focused a great deal of their attention on studies of chemical intermediates in flames (an aspect of flame study to which Neyman [2] has contributed as well). Avramenko alone [4] and then in conjunction with Lorentso [5] are credited with having developed and improved a spectroscopic method for the determination of constants of the rates of reactions of free hydroxyls with other radicals and molecules.

With the use of this method and an arrangement described earlier by Avramenko [4], the [illegible] scientists have succeeded in determining the constants of the rates of the following reactions of free hydroxyls with formaldehyde and acetaldehyde:

$$OH + H_2CO = H_2O + \begin{cases} HCO \rightarrow H + CO \\ \text{or} \\ H + CO \end{cases} \quad \text{and} \quad OH + CH_3CHO = H_2O + \begin{cases} CH_3-C^{\overset{O}{\diagdown}} \\ \text{or} \\ -CH_2-C^{\overset{O}{\diagdown}}_{H} \end{cases}$$

Values in separate tables compiled for each reaction (the rate operation of the jet, the average concentration of the aldehyde, the concentration of hydroxyl in the jet in the absence of material undergoing combustion, the concentration of hydroxyls in the burning jet, and the constant of the reaction rate) were derived from a formula and by methods also mentioned previously by the authors [4-5].

For the initial reaction, the formaldehyde was prepared immediately prior to the experiment by heating paraformaldehyde and

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then cooling the gas with liquid air to prevent polymerization. The reaction itself was not influenced by the temperature in the range from 100° to 250°C, a fact which was attributed to the very small energy of activation (not exceeding 1000 calories), and the researchers claimed that the occurrence of a trimolecular reaction in this case is very unlikely.

The acetaldehyde was obtained by depolymerization of paraldehyde and stored in ampoules. Then, from a graph expressing the relation of lg $\frac{K_{CH_3CHO}}{T}$ and 1/T, E=4000 calories/mole was derived as the energy of activation.

Therefore, from absolute values of the constants for known energies of activation, (Avramenko and Lorentso were able to derive) the pre-exponential functions and subsequently the constants themselves:

$$K_{HCOH} = 10^{-14}\sqrt{T}\, e^{-500/RT} \text{ sec.}^{-1}\text{ cm}^3, \text{ and}$$

$$K_{CH_3CHO} = 2.5\cdot 10^{-12}\sqrt{T}\, e^{-4000/RT} \text{ sec.}^{-1}\text{ cm}^3.$$

Investigations of the type just summarized (considered to be) are of value, since as pointed out by Zel'dovich and I. Ya. Shlyapintokh in the former's most recent analysis of explosive gas mixtures in percussion waves[6], additional information is badly needed on reaction kinetics at high temperatures for clarification of the behavior of flames, the part played by the diffusion of active centers in the mechanism of chemical reactions, and for an understanding of the relationship between normal and spin detonations. They feel that only the oxidation of hydrogen has been investigated to an adequate extent at present.

~~In this line, another contribution (8) in the field of catalysis~~

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and with a possible bearing on the reaction kinetics of flame and combustion phenomena has been made recently by M. V. Polyakov, Director of the Kinetics and Catalysis Division of the Institute of Physical Chemistry imeni Pisarzhevskiy, of the Ukrainian Academy of Sciences in Kiev. Taking the oxidation of ammonia with the aid of a platinum catalyst to nitric oxide as an example, Polyakov demonstrates experimentally that prior to the main reaction there occurs initially a heterogeneous-homogeneous reaction on which the walls of the container have an effect, leading to the defixation of nitrogen and a reduction in the yield of nitric oxide particularly at high temperatures and for lengthy periods of contact. According to the author's own statement, results of such studies are of considerable practical interest not only in connection with reactions which are properly speaking catalytic.

[redacted] A condensation of the paper by Zel'dovich and Simonov (1) is given below:

INTRODUCTION

These authors begin with a resume of what has been done so far in the study of spark ignition of explosive gas mixtures (7-9) [redacted], and then state that in their opinion it was necessary to investigate CO / O_2 mixtures, the combustion of which has been studied also by G. A. Barskiy (10) [redacted].

The properties of these mixtures (particularly the rate of combustion) were greatly varied by adding to them different amounts of hydrogen. The pressure of the explosive mixture was also widely varied.

By passing the spark through non-inflammable gases (ammonia, nitrous oxide), the authors determined the heat energy actually liberated in the gas in the spark gap (under the conditions of their experiment) in relation to the capacity of the condenser

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and the pressure of the gas. The main experiments, however, were conducted by passing the spark through flammable mixtures, beginning with a discharge at a small capacity insufficient to produce ignition and gradually increasing the capacity until they were able to detect that minimum capacity (or heat energy) which was sufficient for igniting the mixture. Given the heat energy of the spark it was also possible to calculate the temperature changes in their spacial and time extent during the distribution of a given quantity of heat in an inert gas.

For the theoretical solution of the problem of ignition it may be necessary, according to Zel'dovich and Simonov, to solve the problem of the propagation of heat in a medium in which the combustion may proceed with the liberation of heat. (They state that) the initial rate of the reaction depends chiefly on the temperature, but that in the zone of high temperature in the flame or in a spark the reaction terminates with the consumption of the reacting substance (fuel or oxygen). (The latter then that) description of ignition thus requires both an equation for the heat energy and another for the thermal conductivity, as well as an equation which determines the concentration of at least one of the reacting substances. ~~is also necessary.~~ The change in this concentration will depend on the expenditure of the substance during combustion and on the diffusion of the substance. The solution of such a system of equations should, they feel, give an absolute value of heat necessary for ignition in relation to the thermal properties of the mixture and the kinetics of the reaction.

Furthermore, (the authors say that) since at the present time there are no direct measurements of the kinetics of combustion at 10^{-3} to 10^{-5} seconds

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and at 2000-3000°K ~~which characterize~~ the prediction of reaction kinetics on the basis of the measurement of the velocity of flame is urgently required. They would relate the energies required for ignition ~~must be related~~ to the rate of flame propagation, on the one hand, and to the thermal properties of the mixture, on the other hand.

Theoretical Relationship Between the Energy of Ignition and the Velocity of the Flame.

The energy of ignition E and the velocity of the flame may be related by means of the theory of dimensions. The quantity of heat can be expressed as a derivative of the specific heat (per unit of mass), the density of the mixture, and the magnitude of the heated volume. A given explosive mixture is characterized by its heat content Q. Units of density can be expressed either in terms of the initial density of the explosive mixture ρ_u, or the density of the products of combustion ρ_b. Lastly, the volume is represented by δ^3, the cube of the width of the zone of propagation of the flame. Therefore, the authors arrive at the equation:

$$E = Q\rho\delta^3 \text{ const} = C_p(T_b - T_u)\rho\delta^3 \text{ const} \quad (1)$$

($Q = \int_{T_u}^{T_b} C_p dT$, where T_b is the combustion temperature and T_u, the initial temperature.) Such a relationship as (1) applies to conditions where the quantity of heat during ignition is sufficient to heat up to the combustion temperature a spherical volume whose radius is the width of the zone of flame propagation.

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The authors specify heating to the combustion temperature, rather than just to any ignition temperature, in line with the theory of propagation of a flame (2-13) in which it was explained that in typical cases the chemical reaction takes place mainly at a temperature close to T_b.

The width of the zone δ is not an exact physical concept: the temperature decreases only gradually as the distance from the flame increases, and the reaction in the products of combustion tapers off only gradually with increasing distance from the flame. It follows from the theory that the reaction proceeds in a narrower zone than the heated zone itself, the latter being found by determining the temperature distribution originating in the gas in front of a surface of known temperature T_b. This surface, which is the reaction zone, moves at a rate equal to the rate of propagation of the flame u. (This problem was first considered by Mikhel'son (13) .)

The distance penetrated by noticeable heating, is expressed by:

$$\delta = \frac{\lambda}{u \rho c_p}, \qquad (2)$$

where λ is the thermal conductivity of the gas. Substituting this value into (1) results in:

$$E = Q \frac{\lambda^2}{u^3 \rho^2 c_p^2} \text{ const} \qquad (3)$$

The conditions of ignition depend essentially on the rate of

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the reaction, because the value δ in equation (1) depends on the rate of the reaction. However, with (2) and (3) the authors succeeded in expressing δ and E in terms of an experimentally observed value for the velocity of the flame.

For explosive mixtures of similar composition, the following equation was arrived at regarding the effect of the pressure of the mixture on conditions obtaining at the moment of ignition and producing the ignition: (λ and C_p do not depend on the pressure and ρ is proportional to the pressure.)

$$E = \text{const} \frac{Q}{u^3 \rho^2} \tag{4}$$

Experimental verification of (1), (2), (3), and (4) and determination of the dimensionless constant appearing in them (which cannot be theoretically determined) was the purpose of this study, according to the authors who then proceed, on the basis of a previous article by Zel'dovich (14), to derive the following expression for the limiting diameter of narrow pipes through which a flame can be propagated with respect to the velocity of the flame:

$$\frac{d u C_p \rho}{\lambda} = \text{const} \tag{5}$$

And with the aid of (2) it is deduced that:

$$d = \delta \, \text{const} \tag{6}$$

Investigations (15) by Lewis and Elbe are cited in confirmation of results obtained in several cases in this section, but the Russian authors also take up some space criticizing the Americans for errors and mention that the latter did not measure the spark coefficient (a technique which Zel'dovich and Simonov take up

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in detail in a later section of this article).

Arrangement of the Experiment

The spark was produced from high voltage apparatus. The high voltage itself is produced on the terminals of a step-up transformer and then rectified by a KR-110 kenotron. (The rectification is half-wave.) The rectified current then, is applied to plates of the filter condenser (2 mfd.) From the filter condenser the rectified and smoothed out high voltage is fed to the cathode of the KR-110 kenotron which serves as a high-ohmic resistance. The kenotron cathode is fed from a separate step-down transformer which has a rheostat in the circuit of the primary winding. By changing the current strength in the primary winding of the step-down transformer with the aid of the rheostat, it was possible thereby to change the incandescence of the kenotron cathode, and correspondingly, its anode current. From the kenotron anode, the high voltage is fed onto the plates of a removable condenser which is discharged in the spark gap of the reaction chamber when the breakdown voltage is reached. In parallel with the spark gap there is a 5-KV high-ohmic (internal resistance of 25 megohms) electromagnetic volt-meter, by means of which the spark-over voltage is measured. (The current is selected so that it can be easily observed as the voltage is gradually increased; in this way the maximum voltage attained just before the spark-over can be recorded. The maximum voltage obtained is regulated with a rheostat included in the circuit of the primary winding of the step-up transformer. The capacity of the removable condensers is measured on a capacitance bridge with a good degree of accuracy.

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The gas apparatus itself consists of a flask for the compounding and storage of the explosive mixture, a pressure gauge, an explosion chamber with a spark gap, a McLeod gauge, and two traps cooled as needed with liquid oxygen. One of the traps precedes the flask in arrangement and the second comes after the McLeod gauge. The explosion chamber consists of a heavy-walled glass flask with sealed-in small wires. On the ends of the wires are small balls (1-1.5 mm in diameter); and between them is the spark gap, about 0.4 mm in length.

In experiments with mixtures of CO and O_2, the explosion chamber had a volume of 57 cm^3; in experiments with mixtures of H_2 / air and with H_2 / O_2, the size of the chamber was about 2 cm^3.

The explosive gas mixture was allowed to flow at the desired pressure into the previously evacuated (up to 10^{-5} mm Hg) explosion chamber; then the spark apparatus was connected by means of cut-outs to the rheostat in the circuit of the first winding of the step-up transformer. The rheostat was gradually eliminated up to the moment of the spark. In this way the voltage was gradually increased across the spark gap. (The voltage was continuously observed on the volt-meter.) Separate experiments were used to determine both the voltage at the moment of discharge and whether ignition occurred or did not occur.

Zel'dovich and Simonov calculated the heat energy liberated in the gas as a result of the discharge for each experiment performed, according to the capacity C and the measured voltage V:

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Results of Experiments on Spark Ignition

These authors point out the difficulty of reproducing the breakdown voltage for a given distance between electrodes, and in fact such variations occur as almost to render this method erroneous. (Under equal external conditions, the mixture is sometimes ignited and sometimes not.) However, because the breakdown voltage was measured directly for each spark, they showed that under equal conditions those sparks ignited for which V (and therefore, E_e and E_t) were large.

For example, in the mixture of 1.25% H_2 / 87.55% CO / 11.2% O_2, with P = 380 mm, C = 0.0026 mfd, V = 2650 V, and $E_e = 9.1 \cdot 10^{-3}$ J, ignition did occur. For V = 2000 V and $E_e = 5.2 \cdot 10^{-3}$ J, ignition did not occur.

Experiments were conducted with varying amounts of H_2 replacing CO in the basic mixture 88.8% CO / 11.2 O_2 and also with varying pressures under these conditions; the results were depicted on four graphs. Along the X axis in the first graph was represented the percentage of H_2 at a constant pressure of 380 mm; in the latter three graphs, the pressure of a mixture of a given composition (2.5% H_2, 1% H_2, and 0.3% H_2, respectively) was shown along the X axis. In all four cases, the Y axis was used to measure the electrical energy E. All graphs are logarithmic with respect to both the X and the Y axis.

In these four graphs, the region of ignition is shown to the right and to the top, and a smooth curve separates this region from the non-ignition region to the left and to the bottom, making

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it possible to predict (within limitations, since the data are fragmentary, as Zel'dovich and Simonov themselves point out) the minimum energy required to ignite a given mixture.

A few experiments using different electrode intervals with the same mixture led to the results that the E_e required for ignition may be decreased approximately 35% during an increase of the interval of from 0.4 to 1.1-212 mm.

The velocity of Flame in the 88.8% CO / 11.2% O_2 Mixture

Here, calculations made previously (10) by Barskiy with the use of a rotating film are incorporated, and an interpolational equation is derived from his data:

$$u_b = 81\sqrt{H_2}\sqrt{P} \qquad (9)$$

H_2 is given in percent; P in mm; and u_b (representing the rate of the flame with regard to the quiescent products of combustion) in cm/sec. Two tables are then presented to show the close conformity of u_b obtained experimentally by Barskiy and u_b calculated with equation (9).

Comparison of Experimental Results with the Theory

As stated earlier, the authors anticipated that the minimum energy of the spark required for ignition may be represented as that energy which is necessary to heat a spherical volume to the combustion temperature. The radius R of this sphere is proportional to δ, the width of the heated zone in a stationary flame. Comparing theory with experiment, this relationship is seen to be valid.

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Calculation of R is performed according to the equation:

$$\frac{4}{3}\pi R^3 \cdot \rho_b' \cdot C_p (T_b - T_0) = \frac{4}{3}\pi R^3 Q \cdot \rho_b' = E_z = E_e \cdot \eta \quad (10)$$

where ρ_b' is the density of the mixture at a given pressure and combustion temperature T_b, and Q is the specific (per gram) heat of combustion. For the basic mixture of $CO + O_2$ investigated, Q is equal to 15.1 kilocarlories/mole or 2230 J/g. According to thermodynamic calculation, T_b is 2100°K.

The product $Q \cdot \rho_b$ does not depend on the quantity of H_2 in the mixture, and the following equation was obtained: (R in cm, E_e in J, and P in mm)

$$R = \sqrt[3]{\frac{3}{4\pi}\frac{E_e \cdot \eta}{Q \cdot \rho_b}} = 7.8\sqrt[3]{\frac{E_e \cdot \eta}{P}} \quad (11)$$

In calculating δ it was assumed that the chief role in the flame is played by the range heated close to the combustion temperature T_b. (In this range, the composition of the gas is also close to the composition of the products of combustion. (11-12)

On the basis of this assumption a further relationship was evolved:

$$\delta = \frac{\lambda_b}{\rho_b C_{pb} u_b} \quad (12)$$

In calculating the composition of the products of combustion, it was assumed for practical purposes that the H_2 did not react with O_2 in the presence of the excess of CO.

The product $\rho_b u_b$ represents the quantity of a substance burned in a unit of time on a 1 cm^2 area of flame surface ($\rho_b \cdot u_b$ and

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$u_n \cdot u_n$ are identical.)

Conversion to combustion temperature was done by Zel'dovich and Simonov with Sutherland's equation, assuming C = 150. For mixtures containing an excess of CO, ($T_b = 2100^\circ K$) λ_b varied only slightly ($22 \cdot 10^{-5}$ on the average.)

The thermal conductivity of the mixture in its original state differs only slightly from that of the products. A greater discrepancy occurs for mixtures of H_2 / air and mixtures of CH_4 / O_2 (the latter investigated by Lewis and von Elbe).

Results of all experiments are summarized in a general graph, lg R mm lying along the ordinate axis and lg mm along the X axis. Despite the sparsity of data, the regularity of increase in R as increases is shown. When R and are large, it can be approximately stated that:

$$R = 3 \cdot , \qquad (13)$$

which leads to the equation:

$$E_e = \frac{12\pi}{\eta} \cdot Q \frac{\lambda_b^3}{u_b^3 C_{pb} \rho_b^2} \qquad (14)$$

The graph shows that in very lean hydrogen (slowly combustible) mixtures at very low pressures, the size of the spark is very large: R up to 6 mm at 1% H_2 and 50 mm Hg. Because the breakdown voltage of a gas at low pressure is small, a large capacity up to 2 mfd is required. It is very possible that in a number of cases the measured limits for the propagation of a flame at low pressures are actually

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the limits of the ignition and that a greater intensification of the ignition may cause displacement of these limits. For observation of the propagation of a flame, it is necessary to use sufficiently large containers (with the radius many times greater than R).

The curves on the graph are in conformity with experiments, but divergencies do occur when R and ' are small, when ' is less than and R is comparable with the dimension of the spark gap.

Photographic Study of Ignition

Zel'dovich and Simonov prepared some photographs of sparks which produced ignition, and also of some which did not:

The photographs were made with a mixture of 88.7% CO / 11% O_2 / 0.3% H_2 (giving the least u_b and the greatest ' at a given pressure) in the P range of from 100 to 400 mm. A rotating film with a speed of about 5 m/sec was used to make pictures 1/5 actual size. The flask was housed in a screen with an aperture of about 4 mm. (In several experiments with great spark energy, this aperture was decreased to 0.5 mm.) The electrodes were perpendicular to the aperture and the objective to the axis of the objective lens. The observed radius of the luminous sphere at a spark energy sufficient to ignite the mixture was found to be on the average 1.3 times greater than the calculated value for R. Evidently, the liberation of chemical energy in the spark plays a role in the reaction because, in the chilled products of combustion in which the gas of equal density and with an equal spark capacity, the radius of the luminous sphere is 1.8 times smaller than the corresponding radius in the burning mixture, or 1.4 times less than the calculated value for R.

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An interesting initial decrease was observed for R even in cases where ignition occurred; evidently, in this initial period, the loss of energy (by radiation) of the incandescent gas in the middle of the volume heated by the spark is greater than the reaction energy evolved on the surface of the spark. That is why in the initial period this cooling leads to compression, and why the heated gas moves to the center carrying the flame with it. Only in the following stage does propagation of the flame begin at an approximately constant rate.

In no case was there observed a complete interruption in the luminescence. However, between the spark discharge and the uniform propagation of the flame, there is an interval of time in which the sphere of the flame is small, and the illuminated layer is thin, and the intensity of luminescence is small also. The Russians suggested that with sufficiently sensitive photographic materials, a conclusion may be reached with regard to the nature of this induction period, admitting they had not fully explained the photographs and regretting the difficulty of obtaining sharp pictures of a weak flame in the strong light of a powerful spark.

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List of Symbols in the Order of their Introduction into the Text

r	-	radius of a sphere which can be heated with a spark to the combustion temperature (in cm)
	-	width of the heated zone in the flame
E	-	energy of ignition of a flame
	-	heat of heat of a given explosive mixture (per gram)
ρ_u	-	initial density of the explosive mixture
ρ_b	-	density of the products of combustion
V^3	-	volume of the heated zone of the flame
C_P	-	molecular heat at constant pressure
T_b	-	combustion temperature
T_u	-	initial temperature
u	-	rate of propagation of a flame
λ	-	thermal conductivity of a gas
d	-	limiting diameter of a narrow pipe through which a flame can be propagated
C	-	capacity (in farads)
V	-	voltage (in volts)
E_t	-	heat energy liberated in the gas as a result of an electrical discharge (in joules)
η	-	spark efficiency (in %)
E_e	-	electrical energy built up in the condenser previous to the discharge
C	-	capacity (in mfd)
P	-	pressure (in mm)
j or J	-	joules
u_b	-	rate of flame with regard to the quiescent products of combustion (in cm/sec)
ρ_b'	-	density of a mixture at a given pressure

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(E_t in joules, C in farads, and V in volts)

$$E_t = \frac{\eta}{2} C V^2 \qquad (7)$$

η is the spark efficiency, and is equal to E_t/E_e, the heat energy released in the gas over the electrical energy in the condenser previously to the discharge, $E_e = CV^2/2$.

Determination of the Spark Efficiency

To determine η, a series of sparks (from 10 to 2000) was passed through ammonia gas in the reaction chamber. Following this, the flask was connected with the trap immersed in liquid oxygen, and the pressure of the N_2 and the H_2 was measured with the McLeod gauge. Making definite assumptions concerning the energy required to split up ammonia, the authors solved η for different conditions. Thus, the minimum spark energy required to ignite a mixture of a given composition at a given pressure was determined. Experimental data here obtained make it possible to construct curves for the relationship of this minimum energy to the composition of the mixture or (at a constant composition) to the pressure. Detailed tabular results of these experiments are then given, and the following general equation is derived:

$$\eta = 3.3\, C^{-0.29} \qquad (8)$$

where η is in percent and C in microfarsdays. The equation applies to a pressure of 380 mm.

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